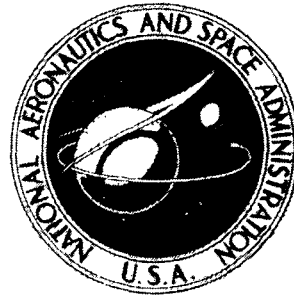


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IRRADIATION TEST OF TUNGSTEN CLAD
URANIUM CARBIDE - ZIRCONIUM CARBIDE
((U,Zr)C) SPECIMENS FOR THERMIONIC
REACTOR APPLICATION AT CONDITIONS
CONDUCTIVE TO LONG-TERM PERFORMANCE

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SUMMARY

Uranium carbide fueled, thermionic-emitter configurations were encapsulated and irradiated in the NASA Plum Brook Reactor. This report discusses two such irradiations designated as V-2E and V-2F. The V-2E capsule contained a specimen clad with fluoride-derived chemically vapor deposited (CVD) tungsten. The other capsule, V-2F, used a duplex clad specimen consisting of chloride-derived on fluoride-derived CVD tungsten. Both fuel pins were 16 millimeters (0.631 in.) in diameter and contained a 45.7-millimeter (1.8-in.) length of fuel.

Irradiation of both capsules began at design operating conditions of 1820 K maximum clad (emitter) surface temperature and 170 watts per cubic centimeter fission heat generation. Shortly after test startup the V-2F capsule exhibited a change in heat-transfer characteristics necessitating operation at a fuel power density of 300 watts per cubic centimeter in order to maintain the design clad temperature. After 1483 hours the V-2F irradiation was terminated because of fission gas leakage. A neutron radiograph showed water in the capsule, possible failure of a containment joint, but no change in fuel or clad geometry. Results of a postirradiation examination are presented.

Irradiation of the V-2E capsule continued to 5957 hours when a similar change in heat transfer necessitated an operating power density of 250 watts per cubic centimeter in order to maintain design clad temperature. After another 500 hours of irradiation at these conditions, the test was terminated because of the cessation of nuclear programs at the NASA-Lewis Research Center.

Fuel thermocouple performance data from several capsule irradiations on this program are summarized.

INTRODUCTION

A program to evaluate refractory-metal-clad, uranium carbide - zirconium carbide ((U, Zr)C) fuel as a candidate emitter-fuel combination for a nuclear thermionic space-power reactor began at NASA about 12 years ago (refs. 1 to 11). One part of this program concerned the irradiation behavior of clad-fuel specimens that simulated the geometry of a fuel element for such a reactor. The design requirements were severe, for example, clad surface temperature, 1800 to 2050 K; fuel burnup, 1.5×10^{20} to 3×10^{20} fissions per cubic centimeter; and cladding dimensional stability, within 3 percent on the diameter. These conditions must be met during a lifetime of 20 000 to 40 000 hours. No fuel-clad candidate has successfully achieved these conditions simultaneously, and especially lacking has been a long-life experiment. According to reference 12 the longest thermionic irradiation was with a uranium dioxide (UO_2)-fueled, tungsten-clad emitter in a two-cell thermionic fuel element; it ran 12 500 hours before the test was terminated because of low power output.

The longest irradiation reported for a carbide-fueled emitter configuration was described in references 13 and 14. In that experiment the clad-fuel specimen in one capsule achieved a burnup of 3.0×10^{20} fissions per cubic centimeter of fuel in 11 090 hours at a maximum clad temperature of 1930 K; however, the tungsten cladding suffered severe localized dimensional changes (ref. 15). After reviewing these results, it was decided that another two-capsule experiment should be conducted with the goal of achieving a very long-term irradiation. To this end, the following conditions were specified:

(1) A maximum clad temperature of 1820 K. This represents a 100 K reduction from the reference 13 test.

(2) A cladding thickness of 1.5 millimeters (0.06 in.). This is a 50 percent increase over the reference 13 test.

(3) A fuel pellet surface area to volume ratio A/V value of 7.17 square centimeters per cubic centimeter. This is a higher value than any used in the reference 13 test.

(4) Periodic capsule rotation to reduce local burnup peaks and attendant cladding deformation which occurred on a previous test (ref. 15).

These conditions were selected to ease the mechanical stress on the cladding and thereby reduce the creep rate. Moreover, it was expected that the lower temperature would not only reduce the extent of fuel-clad interaction but also decrease the diffusion of fuel components through the clad, which has been shown (ref. 11) to deleteriously affect power output from a cesiated thermionic converter.

The performance of these clad-fuel specimens in the reactor environment is described in this report. The termination of nuclear power and propulsion programs at the NASA-Lewis Research Center limited the longest lived capsule test to 6457 hours.

EXPERIMENT APPARATUS

General

Except for a few differences, the components that made up the assembly of the two capsules in this experiment, hereinafter designated V-2E and V-2F, were essentially identical to those described for capsules V-2C and V-2D in references 7 and 13. A longitudinal section through a capsule is shown in figure 1. It was observed during irradiation of capsules V-2C and V-2D (refs. 13 and 14) that leaks occurred between the capsule containments, that is, fuel, primary, and secondary. This leakage caused mixing of the three gases, argon, neon, and helium, in the respective containments and required a substantial increase in fission heat generation rate to achieve design fuel temperature. In an attempt to prevent a reoccurrence of this condition, the V-2E and V-2F capsules were designed such that neon could be used in all three containments; the neon pressure was 1.01×10^4 newtons per square meter (0.1 atm) in the fuel containment and was 1.01×10^5 newtons per square meter (1.0 atm) in the primary and secondary containments, all at ambient temperature. The capsule instrumentation, namely, numbers, kinds, and locations of thermocouples as well as a built-in calorimeter with electric heat source, was the same as described in reference 13 for capsules V-2C and V-2D. An additional feature for capsules V-2E and V-2F was the installation of flux wires in the fuel region of the outer containment to measure the integrated thermal and fast flux.

The capsule irradiations took place in the number 2 vertical tube facility (V-2) of the NASA Plum Brook Reactor at Sandusky, Ohio.

Fuel

The nominal fuel composition for both capsules of this experiment was the same as that used in capsule V-2C, namely, a solid solution of 90 mole percent uranium carbide and 10 mole percent zirconium carbide with 4 weight percent tungsten added. The fuel was enriched 30 percent in uranium-235. However, in an attempt to improve the compatibility between the fuel and cladding and therefore reduce the depth of reaction layer buildup on the cladding below that observed on the V-2C specimen (ref. 15), the carbon to uranium ratio C/U value was specified to be between 1.02 and 1.035; this compares to the 1.03 to 1.05 limits specified for the V-2C fuel. The C/U value is computed by assigning one carbon atom to each zirconium atom and one carbon atom to every two tungsten atoms; the remaining carbon atoms form the numerator of the ratio. The actual C/U value for the fuel used herein was measured to be 1.025 for V-2E and 1.022 for V-2F.

The A/V value, or the ratio of fuel pellet surface area available for fission gas release to the volume of the final machined pellet, was specified to be 7.17 square centimeters per cubic centimeter for all pellets. The configuration of a typical fuel pellet is shown in figure 2. The grooves and small holes are the source of surface area for gas release; the volume of fuel removed by their presence was about 10 percent of the pellet volume before those holes and grooves were machined. The 7.17-square-centimeter-per-cubic-centimeter value is about 4 percent higher than the highest value used in the V-2C irradiation. The fraction of fuel cavity volume occupied by fully dense fuel was determined to be 0.69, using data from reference 16.

Cladding

Emission-diffusion tests of different kinds of tungsten cladding on carbide fuels (ref. 10) indicated that duplex (chloride-derived on fluoride-derived) chemically vapor-deposited (CVD) tungsten was superior to fluoride-derived CVD tungsten in suppressing diffusion of fuel components. In addition, the chloride-tungsten layer could be deposited with a uniform, stable surface crystal orientation which possessed a substantially higher vacuum work function than the conventional fluoride-tungsten (ref. 17). The higher vacuum work function surface, in turn, was a superior electron emitter material for an electric power producing cesiated thermionic converter (ref. 18). The tests of references 10, 17, and 18 were conducted out-of-pile, however, and the integrity of the bilayer material in a neutron environment for long periods of time was unknown. Consequently, it was decided that, although the cladding material for V-2E should be the standard fluoride tungsten, the V-2F capsule would use duplex tungsten. The cladding for both of the specimens had an inside diameter of 12.8 millimeters (0.505 in.) and an outside diameter of 16.0 millimeters (0.631 in.). The thickness of the chloride-tungsten layer on the duplex cladding was 0.38 millimeter (0.015 in.).

Additional details on the fabrication, impurity analysis, and metallography of the specimens are provided in reference 16. Before installation in the reactor test facility, the two capsules were subjected to neutron radiographic examination and determined to be acceptable for irradiation.

EXPERIMENTAL RESULTS

The irradiation of capsules V-2E and V-2F began in September 1971. The clad-fuel specimen in each capsule achieved the design operating conditions, namely, maximum clad temperature of 1820 K and fission heat generation rate of 170 watts per cubic centimeter of fuel volume, shortly after reactor startup. After about 2 hours of operation a

malfunction in the automatic data recording system necessitated moving the capsules into a low-neutron-flux-density position until the recording equipment could be operated normally. Upon reinsertion of the capsules to the previously established flux density position for design condition operation, it was observed that, although capsule V-2E behaved normally, capsule V-2F data indicated a clad temperature about 45 K cooler than before. Throughout the remainder of the reactor cycle (13 days), the mechanism that positioned both capsules was operated to maintain the V-2E specimen clad temperature at the design value (1820 K); the V-2F specimen clad temperature decreased continually, reaching a value of 1505 K at the reactor cycle completion.

Inspection of the data from the built-in calorimeter in capsule V-2F indicated a change (increase) in the conductivity of the gas in the primary containment. The likely source of the contaminant gas was the helium used to pressurize the 4.9-meter (16-ft) long flexible tube that contained the instrumentation leads. This helium was maintained at a pressure of 11.2×10^5 newtons per square meter (11 atm) at all times; apparently it had entered the specimen region of the capsule through some defect and intermixed with the neon at 1.01×10^5 -newton-per-square-meter (1-atm) pressure.

It was decided to continue the irradiation of capsule V-2F even though the required fission heat generation rate would be 300 instead of 170 watts per cubic centimeter. Since the higher value was only about 13 percent greater than the fission heat rate of the V-2C capsule specimen (which ran 11 090 hr; ref. 14) no substantial risk to experiment life was expected. The test of both capsules proceeded uneventfully for two additional reactor cycles. In the fourth cycle, however, the clad temperature of the V-2F specimen unexpectedly dropped 50 K below the design temperature. It was postulated that additional helium had leaked into the capsule containments. In an attempt to stabilize the operating conditions, it was decided that the helium should be replaced with neon. Accordingly, during the reactor shutdown period after the fourth irradiation cycle of this experiment (1483 hr of irradiation), the lead-tube gas was bled off. A sampling detected fission gas; furthermore, fission gas was also detected in the reactor cooling water, and its source traced to the V-2F lead tube. Apparently, a small leak had occurred in the flexible portion of the tube. The capsule was removed from the reactor, and the specimen portion was subjected to neutron radiography.

The neutron radiograph showed evidence of water in the secondary containment and also failure of the joint between the tungsten fuel cup (cladding) and the tantalum tube to which it was diffusion bonded (see appendix A, fig. 3). No changes in the clad diameter or fuel geometry could be detected. Although the diffusion bonded joint appeared to have opened, the tungsten cup did not fall to the lowest possible position; in fact the separation appeared to be only about 10^{-4} meter wide. Such a break, however, would permit communication between the fuel containment and the primary containment; fission products entering the primary containment could cause transient effects on the specimen heat transfer and therefore its temperature. Because the likelihood of a successful capsule

repair was small and prohibitively expensive, the capsule was transferred to the hot cell for disassembly and failure analysis. The examination and results are described in appendix A.

The irradiation of capsule V-2E continued at the design values of clad temperature and fission power for about 4500 hours after capsule V-2F was removed from the reactor. At that time the clad temperature began to drop; the temperature decrement amounted to 280 K for design fission power. It was suspected that the lead-tube helium was leaking into the specimen containments similar to the V-2F case previously described. During the subsequent reactor shutdown period the lead-tube helium was purged and replaced with neon. However, subsequent irradiation showed that the gas contamination problem still existed. Since no evidence of fission gas leakage or unexplainable temperatures were present, the irradiation was continued; at the design temperature, however, about 50 percent more fission power was needed. The increased power was still within the area of interest for the intended application.

Capsule operation continued satisfactorily until the total irradiation time reached 6457 hours. At that time the reactor was shut down and further testing was terminated because of a decision to terminate all nuclear programs at the NASA-Lewis Research Center. No hot-laboratory examination of capsule V-2E is planned.

Table I summarizes the important operating parameters as well as the operating hours for the two capsules. A summary of the fuel thermocouple performance for these and previous capsules is presented in appendix B.

CONCLUDING REMARKS

Two capsules, each containing a tungsten-clad carbide-fueled specimen of thermionic reactor fuel-element geometry, were irradiated in the Plum Brook Reactor. The specimen design operating conditions were a maximum clad temperature of 1820 K and a fission power density of 170 watts per cubic centimeter of fuel volume.

One capsule, designated V-2E, operated for 6457 hours and achieved a burnup of 1.54×10^{20} fissions per cubic centimeter of fuel. The other capsule, designated V-2F, operated 1483 hours and attained a fuel burnup of 5.6×10^{19} fissions per cubic centimeter. Internal faults in the capsules led to intermixing of the lead-tube helium with the neon in the capsule containments and to changes in the specimen heat-transfer conditions. As a result, capsule V-2E was operated about 10 percent of its life at 147 percent of the design fission heat rate, and capsule V-2F operated at 176 percent of the design fission heat rate, nearly all of its life.

Capsule V-2E testing was terminated as a result of a decision to stop all nuclear propulsion and power work at the Lewis Research Center. The testing of V-2F was stopped when fission gas was detected leaking from it into the reactor coolant water. A neutron radiograph showed water in the capsule and an apparent fuel containment bond

failure; however, the specimen geometry was unchanged. The capsule was subjected to a brief hot-cell examination before disposal; the information obtained provided an explanation for the necessity to operate the capsule at off-design heat rate to achieve design specimen temperature.

A discussion of the fuel thermocouple performance of these and other ((U, Zr)C) capsules is also presented.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, May 16, 1973,
503-25.

APPENDIX A

CAPSULE V-2F POSTIRRADIATION EXAMINATION

A postirradiation examination of capsule V-2F was made to determine its mode of failure. The following observations had been made before capsule examination:

- (1) Water had entered the outer containment.
- (2) The integrity of the diffusion bond was questionable.
- (3) Fission gas had entered the lead tube and leaked into the reactor cooling water.
- (4) Lead-tube helium had mixed with the neon in the primary containment.

An explanation of all these observations would have required an extensive hot-laboratory examination, which was not feasible because of schedule and funding limitations. Therefore, the examination only involved checking the most accessible leak sites; namely, the outer containment, the lead tube, the potted thermocouple seals, and the upper thermocouple feedthrough. See figure 3 for these locations.

The capsule was placed in a suitable hot cell with the lead tube extending through a hole in the cell wall into an adjacent area. First, the outer containment of the capsule was punctured (No. 1 in fig. 3). Gas pressure at 3.79×10^5 newtons per square meter (40 psig) existed in this containment; however, no detectable fission gas was noted. The containment was vacuum pumped and sealed off; no change in pressure occurred in 24 hours.

Next, the upper end of the lead tube, which was outside the hot cell, was punctured (No. 2 in fig. 3), and a slight positive pressure ($< 1.36 \times 10^5$ N/m² (<5 psig)) was detected. A gas sample from the lead tube, had a krypton-85 activity of 1.33×10^4 disintegrations per second per cubic centimeter. The lead tube was evacuated to remove the radioactive gases; following this, it was pressurized with 8.65×10^5 newtons per square meters (110 psig) of helium. At the same time a vacuum was maintained in the capsule outer containment via puncture number 1 inside the hot cell. There was no change in vacuum or pressure in over 16 hours. The outer containment in the vicinity of the fuel specimen was heated to a temperature that simulated irradiation experiment conditions while maintaining the pressure and vacuum conditions. No change occurred in the lead-tube or outer containment pressure. The pressure differential was then reversed with 8.65×10^5 N/m² (110 psig) helium in the outer containment and a vacuum in the lead tube. Again, there was no change in pressure or vacuum. While at this condition, all weld joints on the outer containment were surveyed with a helium leak detector. There was no leak detected.

At this point it was decided to puncture (No. 3) the primary containment, pressurize it, and check for leaks. The flexible portion of the lead tube was then stripped away so that the potted thermocouple seals were accessible. The seals were visually inspected

and no discrepancies were noted. After sealing puncture number 1, the capsule was pressurized via puncture number 3 and an interesting phenomenon was noted. Helium at 2.75×10^5 newtons per square meter (25 psig) was admitted into the outer and primary containments and then valved off. The indicated pressure dropped and settled out at 2.54×10^5 newtons per square meter (22 psig) in a few seconds. This procedure was repeated resulting in a lesser pressure drop until, after three or four such cycles, there was no pressure drop. It was postulated that an additional volume was slowly filling with helium - this could only be the fuel containment. This indicated the existence of a leak between the primary and fuel containments, possibly through the diffusion bond, which had appeared cracked in the neutron radiograph.

The helium pressure was maintained in the capsule (all three containments) and the 26 potted seals were enclosed in a plastic bag. Helium was detected in the bag after a few hours, and the only leak was found at a fuel thermocouple potted seal. This leak occurred at the junction of the sheath and potted seal body as noted in figure 3.

The fuel thermocouples were sheathed in tungsten - 26-percent rhenium alloy in the higher temperature region and in stainless steel where the temperature was lower. There was a short distance between the sheaths where the insulated wires were exposed to the fuel containment volume. The hot-cell test helium entered the stainless-steel sheath at this point and left through the defective seal in the lead tube. Similarly, during irradiation, fission gas could have followed the same path and entered the flexible lead tube. The sheaths of all other thermocouples and calorimeter heater leads were continued without a break to the region of the potted seals.

It is postulated that, early during irradiation, the lead-tube helium, at higher pressure than the capsule neon gas, was driven through the defective potted seal into the fuel containment. From here the helium subsequently entered the primary containment through the tungsten-tantalum diffusion bond failure and mixed with the neon. The bond failure may have been caused by excessive tensile stresses due to the off-design gas pressure loading.

The fission gas could have diffused back into the lead tube during irradiation after the lead-tube and fuel-containment pressures were equalized. Another possibility was that the fission gas entered the lead tube when it was bled down for fission gas sampling.

Since no leaks were detected in the outer containment and instrument lead tube, it has not been established how the water entered the capsule or how the fission gas leaked out of the lead tube.

APPENDIX B

PERFORMANCE OF FUEL THERMOCOUPLES

The thermocouples used to sense the fuel temperatures in these (U,Zr)C fueled capsules and in those of references 13 and 14 employed tungsten-rhenium alloy wires at the hot junction, which were insulated with beryllium oxide and enclosed in a tungsten-rhenium sheath. Details of the construction are presented in reference 19 and figure 1. Each capsule contained three pairs of fuel thermocouples; one pair was located at each of three axial locations in the fuel (fig. 1). Each thermocouple contained one W-25Re and one W-3Re alloy wire. The wires from a pair of thermocouples were crimped in a tantalum cylinder to form a common hot junction.

Table I presents performance data of these thermocouples in four capsules that were irradiated in the NASA Plum Brook Reactor. Two capsules, V-2C and V-2D (refs. 13 and 14) experienced over 11 000 hours irradiation, and all of their fuel thermocouples failed in that time. The irradiation of capsules V-2E and V-2F of this report was terminated before any thermocouple failure was noted.

It appears that the tungsten-rhenium alloy thermocouples operating at 1950 K and above failed early during the irradiation experiment. Figure 4 shows the performance of the fuel thermocouples plotted as operating temperature against time. A least-squares curve fit to the solid data points is shown; this may be used to estimate the life of similar thermocouples at a specified operating temperature. For example, if the irradiation of V-2E had continued, failure of the thermocouple operating at 1850 K appears likely in 8100 hours. Failure at about 7300 hours would be expected for the V-2F thermocouple operated at 1875 K.

The maximum thermal fluence to which the thermocouples were exposed was 8×10^{19} neutrons per square centimeter. Reference 20 shows no measurable decrease in thermocouple millivolt output in the applicable range of temperatures at this fluence. Therefore, thermocouple failure in these experiments is attributed to temperature effects only.

It is clear from the data shown that the fuel thermocouples would not have provided useful data over a test goal period of 20 000 to 40 000 hours. This event was anticipated in the capsule design when provisions were made to incorporate a number of more reliable chromel-alumel (C/A) thermocouples in the primary and secondary containment walls. Through correlations established early in the irradiation among these C/A thermocouples, the fuel thermocouples, and the thermal power generation, fuel temperatures could be calculated if the fuel thermocouples subsequently failed. The relative complexity of the capsule design was primarily due to the desire to maintain the fuel and, therefore, clad temperature at the design value for a substantial time period.

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TABLE I. - IRRADIATION HISTORY

Parameter	Capsule	
	V-2E	V-2F
Maximum clad temperature, K	1820	1820
Irradiation time, hr	6457	1483
Fuel burnup:		
fission/cm ³	1.54×10^{20}	5.61×10^{19}
atom % U	0.76	0.28
Subcadmium fluence, neutrons/cm ²	4.1×10^{19}	1.6×10^{19}
Average clad heat flux, W/cm ²	38 ^a (56)	70 ^b (40)
Average fuel power density, W/cm ³	170 ^a (250)	300 ^b (170)
Number of thermal cycles:		
Reactor cycles	17	4
Reactor scrams	7	3
Number of capsule rotations ^c	7	4

^aV-2E operated at the higher power, shown in parenthesis, for the last 500 hr of irradiation.

^bV-2F operated at the lower power, shown in parenthesis, for the first 285 hr of irradiation.

^cThe capsules were rotated 1/4 turn, counter clockwise, about their axes during specific reactor cycle shutdown periods.

TABLE II. - HIGH-TEMPERATURE FUEL THERMOCOUPLE PERFORMANCE

Capsule designation	Capsule irradiation time, hr	Flux ($E < 0.1$ eV), neutrons/cm ² · hr	Thermocouple axial location in center of fuel	Thermocouple number	Average operation temperature, K	Time to failure, hr	Failure mode
V-2C	11 089	8.1×10^{15}	Top	1	1670	9 800	Erratic
				2	1670	9 800	Erratic
			Middle	1	1920	9 500	Erratic
				2	1920	7 581	Down drift - 2 hr
			Bottom	1	1970	4 400	Slow down drift - 4 days
				2	1970	4 400	Slow down drift - 4 days
V-2D	12 031	5.3×10^{15}	Top	1	1620	10 000	Erratic and down drift
				2	1620	10 000	Erratic and down drift
			Middle	1	1945	162	Down drift - 1 hr
				2	1945	164	Sudden down drift, then open - 1/2 hr
			Bottom	1	2035	16	Down drift - 1 hr
				2	2035	1	Down drift - 1 hr
V-2E	6 457	6.4×10^{15}	Top	1	1550	(a)	No failure noted
				2	1550		
			Middle	1	1790		
				2	1790		
			Bottom	1	1850		
				2	1850		
V-2F	1 483	10.7×10^{15}	Top	1	1575	(b)	No failure noted
				2	1575		
			Middle	1	1810		
				2	1810		
			Bottom	1	1875		
				2	1875		

^aTest terminated after 6457 hr.^bTest terminated after 1483 hr.

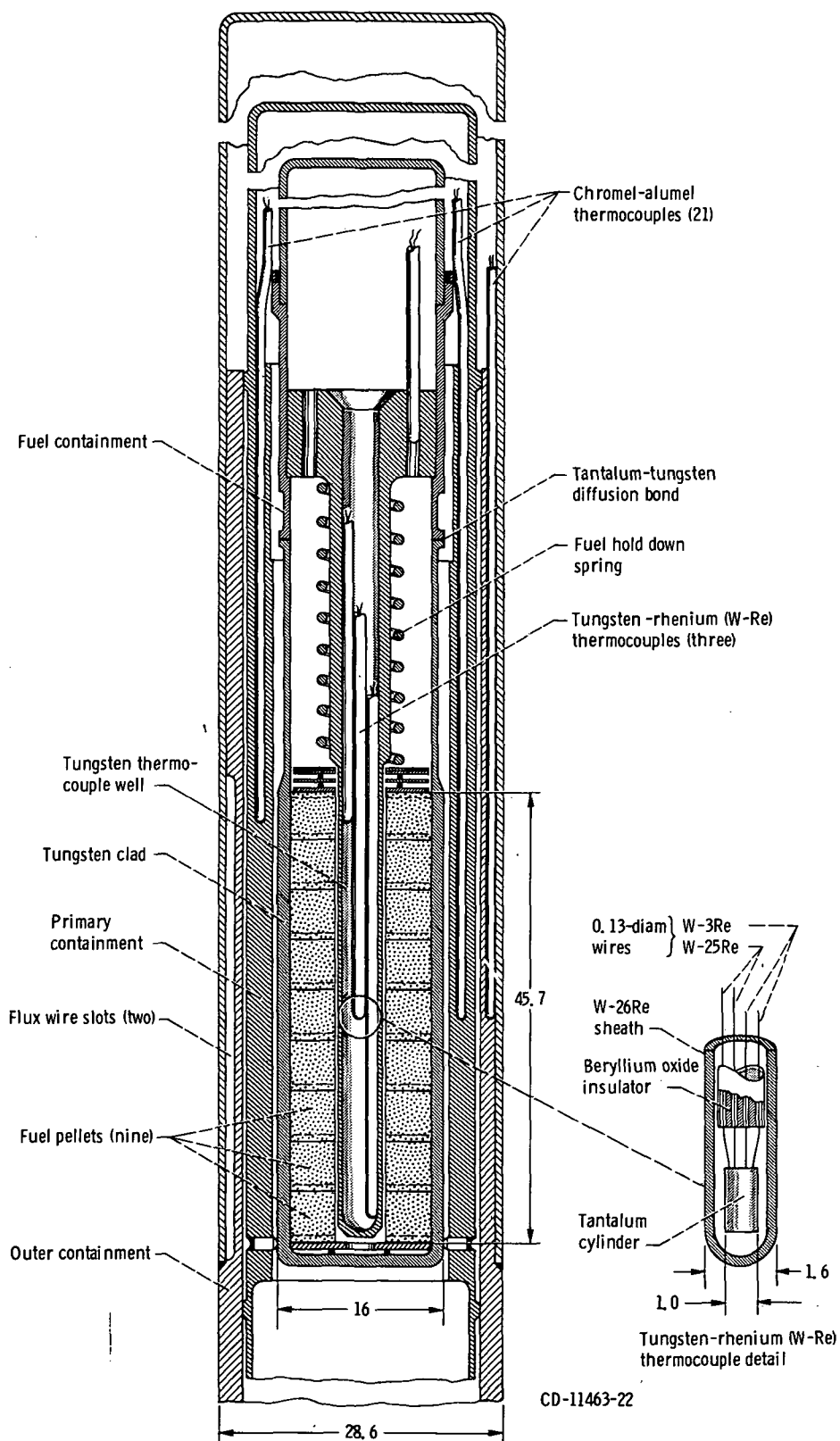


Figure 1. - Longitudinal cross section of capsule in fuel region (dimensions in mm).

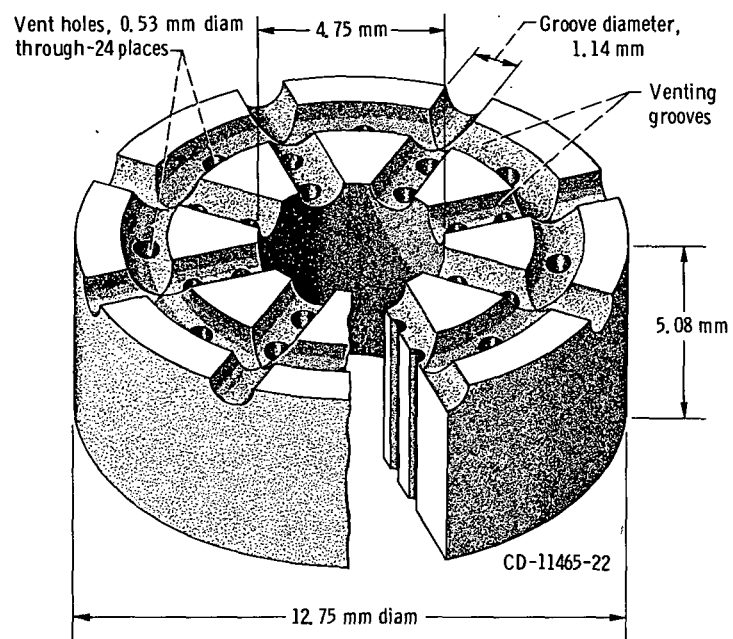


Figure 2. - Fuel pellet configuration for irradiation tests.

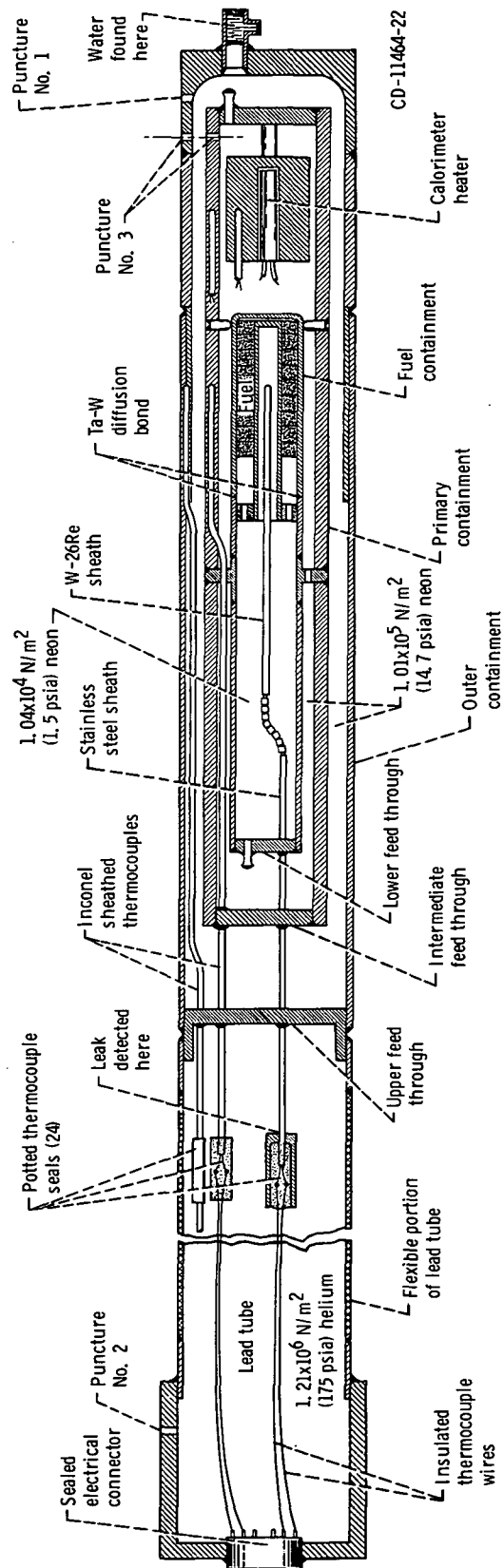


Figure 3. - Schematic longitudinal section of capsule V-2F. Gas pressures were at ambient temperature (20° C) for as-built condition.

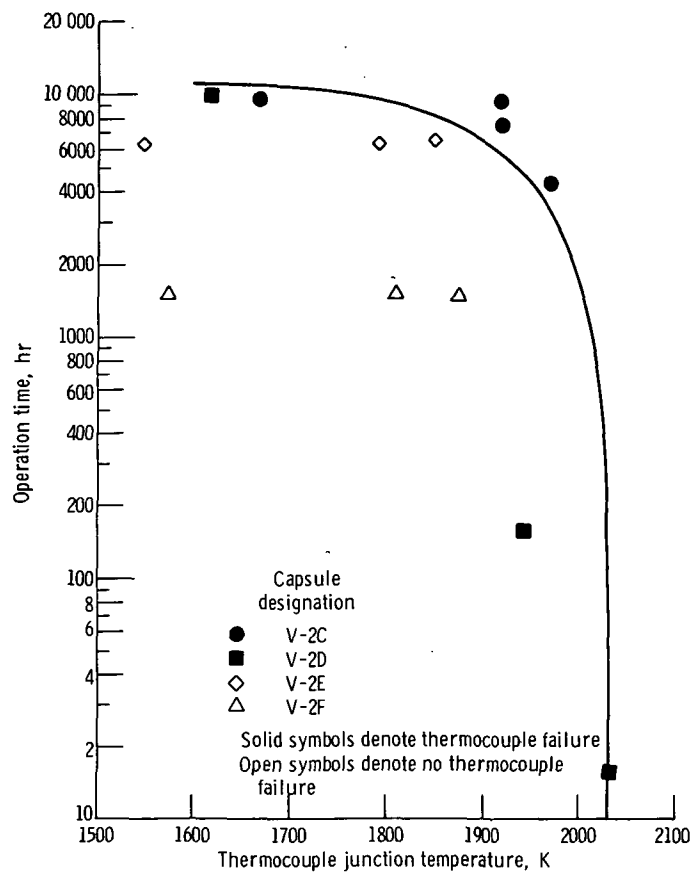


Figure 4. - Fuel thermocouple performance in irradiated capsules.



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